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13. ABSTRACT (Maximum 200 words)

This program has significantly accelerated the construction of a Single Electron Spin Microscope (SESM) – by this time practically completed – and the ideas proposed has been successfully realized. In this final report we emphasize those aspects of the SESM construction, where funding provided under this pilot project made a substantial difference in the planning and design phase of the development even if additional funds were required for the eventual realization, such as: (1) Development and design of stiff cantilever allowing for stable SPM operation and control at sub nanometer sample – tip separations. (2) Broadband detection of the cantilever motion is fed into a computer cluster for 'real time' analyzes; complex distributed software has been developed for that. (3) Satisfactory millimeter wave excitations have been achieved with the developed mm wave power amplifiers and strip-line resonators. At the time of writing this report the SESM construction is practicaly accomplished. While performance refinements and enhancements are continuously added as a result of our ongoing efforts to find optimum-working configuration our current focus is to learn to use the instrument to explore the potentials built into it.

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Accelerated Development of a High Field Single Electron Spin Microscope

G-DAAD19-01-1-0523
Final Report

Karoly Holczer

(1) Forward

"Accelerated Development of a High Field Single Electron Spin Microscope", a pilot project was the first support from DARPA of our effort to construct a microscope with a single electron spin as a probe. This instrument was conceived to provide an atomic scale topographic image of a sample surface, while simultaneously identifying and investigating paramagnetic centers near the surface. SESM and other related methods, generally referred to as Magnetic Resonance Force Microscopy (MRFM) [1], have their common roots in the well-established Atomic Force Microscopy (AFM) [2] techniques. Two features make SESM radically different from other MRFM approaches [3]: the use of a **single-spin as a probe**, and the possible **room temperature operation**.

The prospect of obtaining analytical (i.e. magnetic resonance) information in the form of atomic scale resolution Scanning Probe Microscopy (SPM) holds the promise to revolutionize nanometer scale science and has the potential to transform numerous scientific disciplines. Most significantly, sensing minute structural changes at the individual protein level holds the key for both drug screening and creating bio-molecular sensors and devices. Hundreds of ingenious experiments, individually designed to probe selected molecules, prove this scenario thereby pointing to an urgent need for a *general, enabling tool* in this area. Combining the SESM herein proposed, and the Site Directed Spin Labeling (SDSL) – a genetic engineering technique of attaching a paramagnetic molecule to any specific amino acid – could well be this enabling tool.

To realize such an instrument is clearly a challenge and this program focused to pioneer solutions for the three main technological problems, which were identified as the bottleneck of progress, namely

#1: to create the optimum cantilever (probe) and force detection for SESM,
#2: develop the necessary mm-wave source for magnetic resonance excitation,
#3: the capture of the weak phase incoherent SESM signal – a prelude to handle quantum information.

Today we can report the successful solution of all these problems, although the solution of any one of them fare exceeded the means and the scope of this program, which was meant to pioneer the way one should address them. A year later the MOSAIC program followed effectively complementing the means to enable the realizations tackled by this program. At the time of writing this report the Single Electron Spin Microsope (ESEM) construction is practicaly accomplished: the early focus provided by this project on the critical technological issues allowed us to size up the problems and be completaly efficient with the MOSAIC program.

(2) *Table of Content (N/A)*

(3) *List of Appendixes, Illustrations and Tables*

Figure 1. Thermal noise of an FIB milled cantilever. The spring constant *k* – as determined from the harmonic oscillator fit – is about a factor 10 higher than optimal.

Figure 2. SEM images of a Silex cantilever and the tip. Note the thickness uniformity and the tip sharpness.

Figure 3. The assembled 128 node computer cluster

Figure 4. Partial view of the newly constructed 1.1 Watt 94 GHz transmit-receive module showing the two TRW amplifiers (gray components) combined with Magic T's preceded by the 10 – 84 GHz mixer and other components.

*Figure 5. a***.** Microscope image of the resonator, *b*. transient nutation observed on DPPH (the black spot on the center of the resonator on *a*.) to measure H_1.

(4) Statement of the problem studied

The Single Electron Spin Microscope, SESM, as originally proposed by the PI in 1995 [3], detects the interaction between pairs of spins in close proximity at room temperature. The SESM will provide the following information simultaneously:

- An atomic scale topographic image, as for a standard AFM.
- An image of paramagnetic centers on the surface, correlated with the topographic image.
- Spectroscopic (analytical) information allowing an insight to the chemical nature of the environment of the paramagnetic center.

Our fundamental aim is to achieve single-spin-resolution, force-detected magnetic resonance at room temperature. The SESM concept leads to a number of counterintuitive choices: We are using a *paramagnetic* tip to generate field gradient (instead of a ferromagnetic particle), we use *stiff* cantilevers (instead of the smallest spring constant possible) to combat thermal noise, and we have a *broadband detection* (instead of resonant methods).

To accomplish this, the first groundbreaking support provided under this program. In this final report we emphasize those aspects of the SESM constructed, where funding provided under this pilot project made a substantial difference in the planning and design phase of the development even if additional funds were required for the eventual realization, such as:

- Development and design of stiff cantilever allowing for stable SPM operation and control at sub nanometer sample – tip separations.
- Broadband detection of the cantilever motion is fed into a computer cluster for ‘real time’ analyzes; complex distributed software has been developed for that.
- Satisfactory millimeter wave excitations have been achieved with the developed mm wave power amplifiers and strip-line resonators.

At the time of writing this report the SESM construction is practicaly accomplished. While performance refinements and enhancements are continuously added as a result of our ongoing efforts to find optimum-working configuration our current focus is to learn to use the instrument to explore the potentials built in.

(5) *Summary of the most important results*

Successful operation of an SESM requires a close and stable distance between the sample and the probe. This can only be achieved if the spring constant of the cantilever is larger than the gradient of the attractive forces close to the surface. In practice, this limits the spring constant k to be in the range of 1N/m to 10N/m.

- The resulting force sensitivity loss has to be compensated as much as possible by increasing the deflection sensitivity of the microscope.
- The natural limit to increase sensitivity is the photon shot noise – often talked about but rarely observed.
- The use of stiff cantilevers will reduce/eliminate the thermal noise problem,
- The Q of the free-standing cantilever is irrelevant. Resonant detection is impossible since the motion in the close vicinity to the surface is heavily damped by other interactions.

Our first aim was to develop and create the cantilever responding for these criteria's as well as to adopt our detection system (bandwidth) to it.

Stiff cantilever operation and stable SPM control:

The heart of a force microscope is the probe-cantilever ensemble as it determines the ultimate performance of the microscope. In our case the optical lever detection implemented dictates the dimensions of the cantilever to achieve optimal (shot noise limited) performance. As the Gaussian light spot on the probe is elliptical with dimensions ~16 μm and 22 μm along and transverse to the cantilever, a ~20 μm long and about 25-30μm wide cantilever is required. These parameters allow shot noise limited displacement detection with a noise floor below $5x10^{-15}$ m/sqrt(Hz). Working in lift mode in the close vicinity of the surface sets a lower limit of 1 – 10 N/m to the cantilever force constant. For a silicon monocrystal cantilever with the above dimensions, this translates to a thickness of less than 1μm and a mechanical resonance frequency in the range of 1 - 4 MHz. A significant portion of the budget has been devoted to develop/acquire these kind of custom design cantilevers. Our working conjecture is that any tip is paramagnetic, i.e. paramagnetic centers can be found with high probability at the end of the tip. While nothing specific is known about that, the available controlled etching – oxidizing procedures for Si give a reasonable hope to attempt to control the deft of the paramagnetic centers on the tip, making silicon the preferred material for probe fabrication.

First, we have ordered from *Mikromash* 25- and 12-micrometer long, silicon cantilevers. *Mikromash* attempted two iterations. Both iterations have been unsuccessful to achieve less than 1-micrometer thickness and other geometrical requirements. In order to test the design itself some of the cantilevers have been modified by Focused Ion Beam (FIB) milling. This has been done at the University of Nebraska, Lincoln within the DARPA-MOSAIC program. Figure 1 shows the noise spectra of one of these 'retrofitted' cantilevers. The detection system bandwidth has been increased to 4 MHz. While the perfect correction was not possible (ideally spring constant

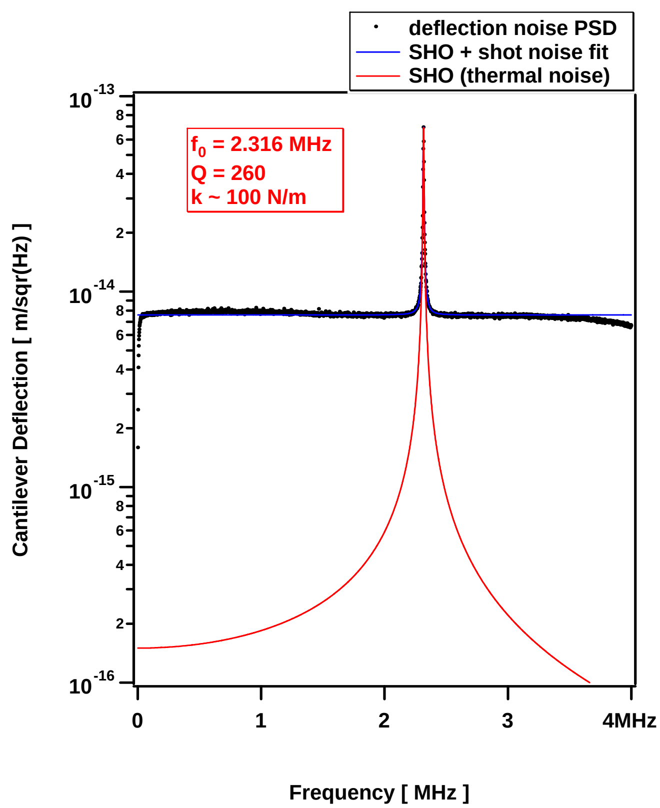


Figure 1. Thermal noise of an FIB milled cantilever. The spring constant k – as determined from the harmonic oscillator fit – is about a factor 10 higher than optimal.

in the 5 – 10 N/m range would be desirable), the result compatible with our expected $2x10^{-15}$ m/sqr(Hz) noise floor for the cantilever design – if properly realized.

While FIB milling is an extremely powerful tool at this stage, the number of cantilevers obtained this way is very limited. Therefore we have continued to search for foundry who would take up on the challenge to fabricate these cantilevers closer to specifications. The second order has been placed with *Silex Micro Systems*. Cantilevers from *Silex* are fortunately falling much closer to the design. A typical cantilever from the Silex batch is shown on Figure 2. Cantilever thickness ranges from 300 to 600 nm over the various parts of the wafer and the oxide sharpened tip height is less than 500 nm. Typical tip radius is less than 20 nm. Several cantilevers (of the order of 50) has been tested and used by now. Their frequency varies in the range of 1.5 – 4 MHz and the spring constant is also in the range of 1 – 50 N/m. All these parameters are in the range we have been aiming for. Using FIB milling on individual probes further improvements or adjustments could be made in the future on the cantilevers presented on Figure 2 as well on other specific designs aiming for high spring constant, compact torsion cantilevers.

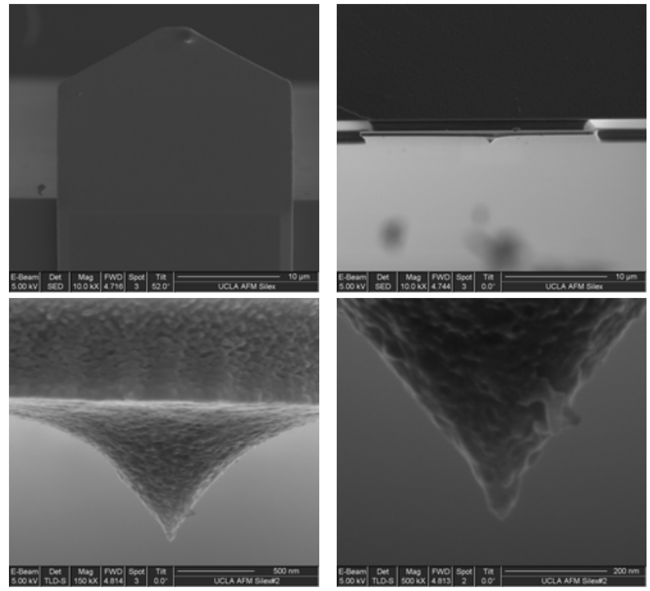


Figure 2. SEM images of a Silex cantilever and the tip. Note the thickness uniformity and the tip sharpness.

These cantilevers satisfy the original specifications aimed for and in practice allow for atomic resolution imaging in contact mode as required for our application. Atomic resolution images using this stiff cantilevers are routinely achieved at 1sec/line scan rate thanks to the exceptional stability of the instrument assembled.

Broadband detection and real time signal analyzes

A 128-node (256 processor) computer cluster has been assembled and tested. Most of the design and planning has been under this program as well as about 20% of the hardware cost has been covered from this funding (complemented from the DARPA-MOSAIC program). Instead of purchasing a complete cluster, we have opted to build it from components, achieving better performance/cost ratio as well as assuring that the a hardware/software architecture is fitting to the nature of the experiment. Data transfer/distribution software development (via Gigabit internet) has been accomplished. Up to 20 MHz digitalization rate the data are broadcast on the network and each node listens and select the data fraction destined to it (typically up 1 – 5 x 10^5 data point) and communicates back the result (the value of a pixel in the image acquired) to the Master computer. This architecture allows for easy change of the data reduction algorithm used by the node and they can be easily programmed and edited using Matlab programming language.

Figure 3. The assembled 128 node computer cluster

Using a modified broad band (up to 10 MHz) detection circuit in conjunction with a specially developed AFM controller (Asylum Research) the cantilever response is digitized at a 10 MHz /32 bit or 20 MHz/16 bit rate synchronous with the millimeter wave excitations. Also synchronized with the motion of the scanner the data is organized in packets representing the image pixels and line by line distributed between the nodes of the cluster, where the values of

each pixel is calculated, than returned to the Master computer for real time display. Several algorithms has been developed and tested in collaboration with the group of Prof. Jose Moura (Carnegie Mellon University, financed by the DARPA-MOSAIC program) to detect the expected phase incoherent small signal.

Millimeter wave power amplifiers and strip-line resonators

A major challenge in the construction of the SESM was to achieve sufficient millimeter wave field strength (more than ~1Gauss) to guarantee well-defined excitations for the qubits (spins) involved. A joint effort with Northrop-Gunman Cooperation (NGC, formerly TRW), Bruker BioSpin Corporation and Radiometer Physik GmbH had been lunched under this program to replace the original 8mW power 94 GHz transmitter – receiver module. Enabled by the DARPA-MOSAIC program we have developed the necessary millimeter wave power amplifiers and strip-line resonators to replace the transmitter – receiver module with an entirely new one, compatible with the rest of the instrumentation. Two MMIC based amplifier units supplied by TRW have been combined using wave-guide Magic T components (see Figure 4) to achieve 1.1 Watts output power. This unit has been fully integrated in the microscope hardware. The present program provided partial financing for various components and the construction of the unit, however the realization required – among others - the execution of a $1.2M subcontract by NGC. This unit was designed to provide about ~ 1 Gauss H_1 field for magnetic resonance excitations using an over coupled Fabry – Perot resonator – a target set as the minimum value where the microscope is expected to become functional.

Further improvements could be expected either by increasing the available power, or changing the resonator structure used and we have pursued both of these options. In the context of the DARPA-MOSAIC program a development and construction of a second unit has been also initiated using a 100-Watts klystron amplifier. This program has largely facilitated and accelerated the design construction of this unit as well. As a more cost effective alternative is to be content with 1.1 Watts and achieve more efficient power to H_1 conversion using different resonator structures. A new strip line resonators have been developed in collaboration with NGC. These resonators have been successfully implemented in the existing Microscope architecture. The resonator serves effectively as a sample holder, while the 94 GHz radiation is coupled to it without mechanical contact, i.e. without compromising the performance of the SPM. Figure 5/a. shows a microscope image the active part of such a resonator with the Silex cantilever placed above it. The

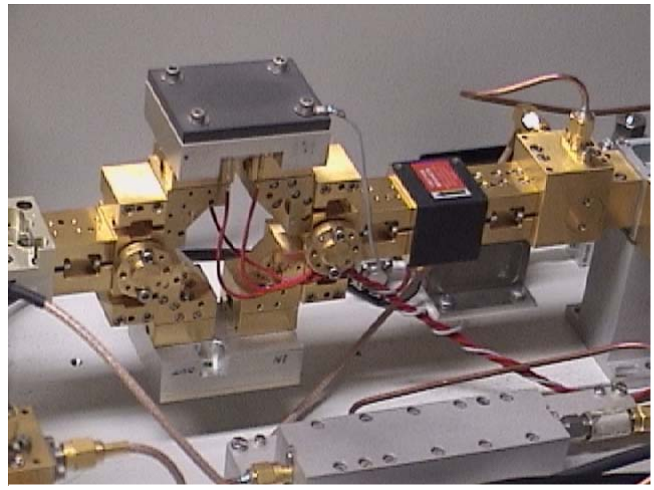
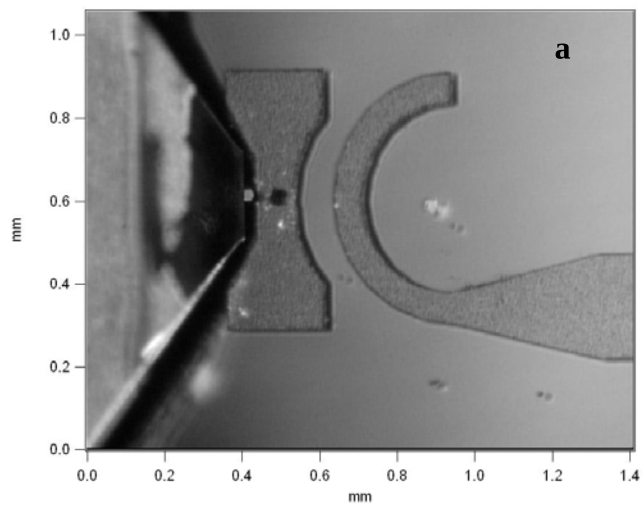


Figure 4; Partial view of the newly constructed 1.1 Watt 94 GHz transmit-receive module showing the two TRW amplifiers (gray components) combined with Magic T's preceded by the 10 – 84 GHz mixer and other components.



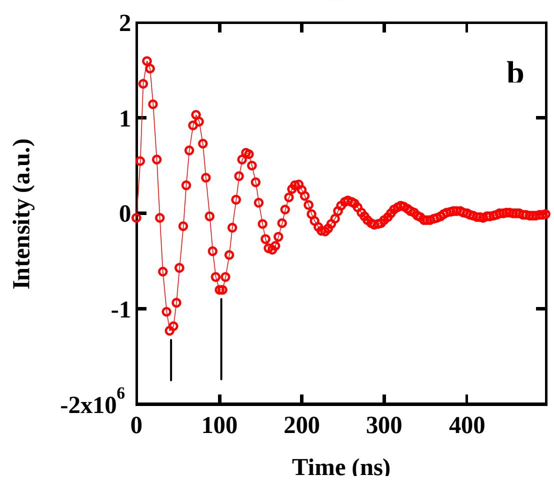


Figure 5. a. Microscope image of the resonator, **b**. transient nutation observed on DPPH (the black spot on the center of the resonator on **a**.) to measure H_1.

black spot in front of the cantilever is a small DPPH crystal, whose observed EPR signal has been used to calibrate the H_1 field. Typically 6 Gauss H_1 has been achieved with this resonator (using 1 Watt power) as shown by the Larmor precession in the rotating frame (called transient nutation) on Figure 5/b. Although the use of this resonator has practical consequences, such as, it limits the useful sample to a about an 0.1 x 0.1 mm^2 area, this inconvenience is largely outweighed by the efficient power – to – H_1 conversion.

Summary

This program has been essential to work out the road map – from the foundry to the UCLA laboratory - to bring about the necessary millimeter wave power, the optimum probe and adequate data analysis power for SESM. Funding provided by this program not only accelerated the development of the constructed SESM, but also contributed significantly to it's realization combined with further resources. In this finally report (delayed pending the outcome of these efforts) we are proud to present the full and successful realization of all the ideas described in the proposal.

(6) Publications:

Due to the nature of the project, no publication has been submitted nor planed on the subjects covered by this project. Several talk has been given however on various review meetings, workshops and conferences by the PI., a list of which follows:

Karoly Holczer: "*Development of a Single Electron Spin Microscope (SESM)*", oral presentation at DARPA MOSAIC Kick-off meeting, April 16th 2002, Washington DC.

Karoly Holczer: "*Single Electron Spin Microscope (SESM)*", oral presentation at CNID Review meeting, August 21, 2002 Los Angeles, CA,

Karoly Holczer: "*Development of a Single Electron Spin Microscope (SESM)*", oral presentation at DARPA MOSAIC Review, October 16, 2002, Los Angeles, CA.

Karoly Holczer: "*Single Electron Spin Microscope (SESM)*", oral presentation at MOSAIC Review meeting, April 29, 2003, Seattle, WA

Karoly Holczer: "*Single Electron Spin Microscope (SESM)*", oral presentation at MOSAIC Review meeting, October 22, 2003, Arlington, VA

Karoly Holczer: "*Single Electron Spin Microscope (SESM)*", oral presentation at CNID Review meeting, August 03, 2004 Santa Barbara, CA

Karoly Holczer: "*Single Electron Spin Microscope (SESM)*", From Solid State to Bio- Physics II, 26/06 – 02/07/2004, Cavtat, Croatia

Karoly Holczer: "*Single Electron Spin Microscope (SESM)*", presentation at AMRI-DARPA. WORKSHOP New Orleans, LU, 2/19-20/2004

(7) Scientific Personal: non

(8) *Report of Inventions*:

"Application of MMIC amplifiers for SESM and high field EPR spectroscopy"
Preliminary list of inventors: K. Holczer, UCLA
R. Lai, NGC
D. Schmalbein & P. Hofer, Bruker BioSpin

"Application of MMIC amplifiers for SESM and high field EPR spectroscopy", a joint UCLA – NGC – Bruker patent, application is pending. The intention is to pave the way to incorporate this technology for a future SESM instrument and also existing EPR spectrometers. The process is pending waiting completion of various tests as well as decisions of our industry partners.

(9) Bibliography

1. Wiesendanger, Roland, "*Scanning Probe Microscopy and Spectroscopy*", Cambridge University Press, (1994)
2. D. Rugar, C. S. Yannoni and J. A. Sidle, "*Mechanical detection of magnetic resonance*" Nature, **360**, 563-566, (1992)
3. K. Holczer, D. Schmalbein and P. Hofer; US Patent #5,619,139, (1997)